This Page Is Inserted by IFW Operations and is not a part of the Official Record

BEST AVAILABLE IMAGES

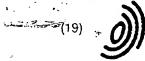
Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

As rescanning documents will not correct images, please do not report the images to the Image Problem Mailbox.



Europäisches Patentamt

European Patent Office

Office européen des brevets



(11) EP 0 753 540 A2

(12)

EUROPEAN PATENT APPLICATION

(43) Date of publication: 15.01.1997 Bulletin 1997/03

(51) Int. Ct.⁶: C08L 69/00, C08K 5/00

(21) Application number: 96111313.1

(22) Date of filing: 12.07.1996

(84) Designated Contracting States: BE DE FR

(30) Priority: 12.07.1995 JP 176248/95 26.07.1995 JP 190657/95 20.09.1995 JP 241546/95 28.11.1995 JP 309211/95 28.11.1995 JP 309212/95 23.02.1996 JP 36344/96 19.03.1996 JP 62615/96

(71) Applicant: MITSUBISHI ENGINEERING-PLASTICS CORPORATION Tokyo 104 (JP) (72) Inventors:

Miya, Shinya,
 c/o Mitsubishi Eng.-Plastics Corp.
 Hiratsuka-shi, Kanagawa-ken (JP)

Kanayama, Satoshi,
 c/o MitsubIshi Eng.-Plast. Corp
 Hiratsuka-shi, Kanagawa-ken (JP)

Shimomai, Ken,
 c/o Mitsubishi Eng.-Plastics Corp.
 Hiratsuka-shi, Kanagawa-ken (JP)

(74) Representative: Hansen, Bernd, Dr. Dipl.-Chem. et al
Hoffmann, Eitle & Partner,
Patentanwälte,
Arabellastrasse 4
81925 München (DE)

(54) Polycarbonate resin composition

(57) A polycarbonate resin composition according to the present invention is suited to be used for medical supplies or appliances and shows a very low yellow discoloration when exposed to an ionizing radiation for sterilization. The polycarbonate resin composition comprises 100 parts by weight of a polycarbonate resin; 0.01 to 5 parts by weight of an aromatic compound containing oxy group or carbonyl group; and 0.01 to 5 parts by weight of at least one compound selected from the group consisting of: (A) a compound containing cyclic acetal group; (B) a sulfone compound; (C) a sulfoxide compound; (D) polyalkylene glycol, an ether of polyalkylene glycol or an ester of polyalkylene glycol; (E) a compound containing sulfonate group; and (F) an aromatic hydrocarbon-aldehyde resin.

BACKGROUND OF THE INVENTION

The present invention relates to a polycarbonate resin composition, and more particularly it relates to a polycarbonate resin composition having an excellent resistance to an ionizing radiation, which exhibits a low discoloration without deterioration in properties thereof when exposed to the ionizing radiation for sterilization.

Polycarbonate resins have been widely utilized in the fields of medical supplies and appliances because the polycarbonate resin have excellent mechanical strength, impact resistance, heat resistance and transparency as well as a high safety. The medical supplies and appliances are generally sterilized upon use. Specifically, the sterilization of these medical supplies and appliances has been carried out by a high-pressure steam sterilization process, an ethyleneoxide gas (EOG) sterilization process, or a sterilization process using an ionizing radiation such as a gamma radiation or an electron beam. Among these sterilization processes, the high-pressure steam sterilization process has disadvantages such as high energy cost and necessity of a drying step after the sterilization. Similarly, the EOG sterilization process has posed such problems as toxicity of ethyleneoxide itself or environmental pollution upon disposal thereof. Under these circumstances, a recent tendency is such that the sterilization process using an ionizing radiation, generally a gamma radiation has been predominately adopted because the sterilization process is relatively inexpensive and can be performed at a low temperature in a dry condition.

However, there also is such a problem that the polycarbonate resin suffers from yellow discoloration when exposed to the ionizing radiation for sterilization thereof. This results in considerable deterioration in value of the products, especially when the polycarbonate resin is applied to the medical supplies or appliances.

As means for solving these problems, there have been proposed a method in which a halogen-containing polycar-bonate resin is used (Japanese Patent Application Laid-open (Kokai) No. 2-55062 (1990)), a method in which a brominated phthalic acid derivative is blended (Japanese Patent Application Laid-open (Kokai) No. 5-179127 (1993)), or the like. However, in these methods, metal corrosion is likely to generate due to the inclusion of halogen in the resin compositions, so that a molding machine or the like therefor are required to be formed from special metal material having a corrosion resistance.

In order to solve the afore-mentioned problems such as yellow discoloration or the like, there have been also proposed a method in which thio-alcohols are used (Japanese Patent Application Laid-open (Kokai) No. 1-229052 (1989)), a method in which thio-ethers are used (Japanese Patent Application Laid-open (Kokai) No. 2-115260 (1990)), a method in which compounds having a mercapto group are used (Japanese Patent Application Laid-open (Kokai) Nos. 2-49058 (1990), 4-36343 (1992) and 6-166807 (1994)), a method in which polyalkylene-oxide and an aromatic compound having a sulfonate group are used in combination (Japanese Patent Application Laid-open (Kokai) No. 5-209120 (1993)), and a method in which compounds having a sulfide group are used (Japanese Patent Application Laid-open (Kokai) No. 6-93192 (1994) and 6-248170 (1994)). However, in these methods, there still exist such problems that the yellow discoloration cannot be sufficiently prevented, or deteriorated environmental conditions occurs due to generation of malodor upon forming, or the like.

Furthermore, in order to solve the afore-mentioned problems, Japanese Patent Application Laid-open publication (Kokai) No. 61-215651 (1994) discloses a method in which boron compounds are used, Japanese Patent Application Laid-open (Kokai) No. 62-135556 (1987) discloses a method in which polyether-polyol or an alkyl ether thereof are used, Japanese Patent Application Laid-open (Kokai) Nos. 64-22959 (1994) and 1-161050 (1989) disclose a method in which polyether-polyol protected by particular terminal groups are used, Japanese Patent Application Laid-open publication (Kokai) Nos. 1-270869 (1989) and 2-13460 (1990)) disclose a method in which polypropylene glycol is used, Japanese Patent Application Laid-open (Kokai) No. 2-232258 (1990) discloses a method in which benzyl alcohol derivatives are used, and Japanese Patent Application Laid-open (Kokai) No. 2-132147 (1990) discloses a method in which itaconic acid, benzaldehyde-dimethylacetal, benzaldehyde-propyleneglycolacetal, etc. are used. However, these methods have also posed such problems that the yellow discoloration cannot be sufficiently prevented or otherwise, if the amounts of the compounds used therein increases to an extent enough in order to prevent the yellow discoloration, the other properties of the polycarbonate resin compositions are adversely affected.

In view of these problems encountered in the prior arts, a polycarbonate resin composition which is capable of exhibiting little deterioration of the inherent properties thereof, little generation of malodor upon forming and little generation of the yellow discoloration upon irradiation of an ionizing radiation for sterilization are strongly demanded.

As a result of the present inventors' intense studies for solving the afore-mentioned problems, it has been found that by blending an oxy group- or a carbonyl group-containing aromatic compound and a specific compound are blended with a polycarbonate resin at specified blending ratios, the obtained polycarbonate resin composition not only maintains inherent properties of polycarbonate resin without deterioration but also prevents generation of yellow discoloration upon sterilization using an ionizing radiation. The present invention has been achieved on the basis of the findings.

SUMMARY OF THE INVENTION

It is an object of the present invention to provide a polycarbonate resin composition which is suited to use for medical supplies or appliances and shows a very low yellow discoloration when exposed to an ionizing radiation for sterilization, while preventing deterioration of inherent properties thereof.

To accomplish the aims, in an aspect of the present invention, there is provided a polycarbonate resin composition comprising 100 parts by weight of a polycarbonate resin; 0.01 to 5 parts by weight of an oxy aromatic compound containing oxy group or carbonyl group; and 0.01 to 5 parts by weight of at least one compound selected from the group consisting of: (A) a compound containing cyclic acetal group; (B) a sulfone compound; (C) a sulfoxide compound; (D) polyalkylene glycol, an ether of polyalkylene glycol or an ester of polyalkylene glycol; (E) a compound containing sulfonate group; and (F) an aromatic hydrocarbon-aldehyde resin.

In a second aspect of the present invention, there is provided a polycarbonate resin composition comprising 100 parts by weight of a polycarbonate resin; 0.01 to 5 parts by weight of an aromatic compound containing oxy group or carbonyl group; 0.01 to 5 parts by weight of a compound containing cyclic acetal group; and 0.01 to 5 % by weight of polyalkylene glycol, an ether of polyalkylene glycol or an ester of polyalkylene glycol.

DETAILED DESCRIPTION OF THE INVENTION:

The present invention is described in detail below.

The polycarbonate resin used in the composition according to the present invention may be homopolymers or copolymers of a linear or branched thermoplastic aromatic polycarbonate prepared by reacting an aromatic dihydroxy compound or a mixture of the aromatic dihydroxy compound and a small amount of a polyhydroxy compound with phosgene or diester of carbonic acid.

As the aromatic dihydroxy compounds, bis(hydroxyaryl)alkanes such as 2, 2-bis(4-hydroxyphenyl)propane (= bisphenol A), 2, 2-bis(3, 5-dibromo-4-hydroxyphenyl)propane (= tetrabromo-bisphenol A), bis(4-hydroxyphenyl)methane, 1, 1-bis(4-hydroxyphenyl)ethane, 2, 2-bis(4-hydroxyphenyl)butane, 2, 2-bis(4-hydroxyphenyl)octane, 2, 2-bis(4-hydroxyphenyl)octane, 2, 2-bis(4-hydroxyphenyl)octane, 2, 2-bis(4-hydroxy-3, 5-dimethylphenyl)propane, 2, 2-bis(3-bromo-4-hydroxyphenyl)propane, 2, 2-bis(3, 5-dichloro-4-hydroxyphenyl)propane, 2, 2-bis(3-cyclohexyl-4-hydroxyphenyl)propane, 1, 1-bis(4-hydroxyphenyl)propane, 2, 2-bis(3-cyclohexyl-4-hydroxyphenyl)propane, 1, 1-bis(4-hydroxyphenyl)-1-phenyl ethane or bis(4-hydroxyphenyl)diphenylmethane; bis(hydroxyaryl)cycloalkanes such as 1, 1-bis(4-hydroxyphenyl)cyclohexane; dihydroxy-diaryl ethers such as 4, 4'-dihydroxy-diphenyl ether or 4, 4'-dihydroxy-3, 3'-dimethyldiphenyl ether; dihydroxy-diaryl sulfides such as 4, 4'-dihydroxy-diphenyl sulfoxide or 4, 4'-dihydroxy-3, 3'-dimethyldiphenyl sulfoxide; dihydroxy-diaryl sulfoxides such as 4, 4'-dihydroxy-diphenyl sulfoxide or 4, 4'-dihydroxy-3, 3'-dimethyldiphenyl sulfoxide; dihydroxy-diaryl sulfoxides such as 4, 4'-dihydroxy-diphenyl sulfoxide or 4, 4'-dihydroxy-3, 3'-dimethyldiphenyl sulfoxide; dihydroxy-diaryl sulfoxides such as 4, 4'-dihydroxy-diphenyl sulfoxide or 4, 4'-dihydroxy-3, 3'-dimethyldiphenyl sulfoxide; dihydroxy-diaryl sulfoxides such as 4, 4'-dihydroxy-diphenyl sulfoxide or 4, 4'-dihydroxy-3, 3'-dimethyldiphenyl sulfoxide; dihydroxy-diaryl sulfoxides such as 4, 4'-dihydroxy-diphenyl sulfoxide or 4, 4'-dihydroxy-3, 3'-dimethyldiphenyl sulfoxide; dihydroxy-diaryl sulfoxides such as 4, 4'-dihydroxy-diphenyl sulfoxide or 4, 4'-dihydroxy-3, 3'-dimethyldiphenyl sulfoxide; dihydroxy-diaryl sulfoxides such as 4, 4'-dihydroxy-diphenyl sulfoxide or 4, 4'-dihydroxy-3, 3'-dimethyldiphenyl sulfoxide; dihydroxy-diaryl sulfoxides such as 4, 4'-dihydroxy-diphenyl sulfoxide; dihydroxy-diaryl sulfoxides such as 4, 4'-dihydroxy-diphenyl s

In addition, branched polycarbonate resins can be obtained by using a polyhydroxy compound such as fluoroglucine, 2, 6-dimethyl-2, 4, 6-tri(4-hydroxyphenyl)-3-heptene, 4, 6-dimethyl-2, 4, 6-tri(4-hydroxyphenyl)-2-heptene, 1, 3, 5-tri(2-hydroxyphenyl)benzole, 1, 1, 1-tri(4-hydroxyphenyl)ethane, 2, 6-bis(2-hydroxy-5-methylbenzyl)-4-methylphenol or a, a', a''-tri(4-hydroxyphenyl)-1, 3, 5-triisopropylbenzene; 3, 3-bis(4-hydroxyaryl)-oxindole (= isatin-bisphenol); 5-chloroisatin-bisphenol; 5, 7-dichloroisatin-bisphenol; 5-bromoisatin-bisphenol; or the like.

In the case where the polycarbonate resin is prepared according to a phosgene method, the reaction may be carried out in the presence of a terminator or a molecular weight modifier. Specific examples of the terminators or molecular weight modifiers may include phenol, p-t-butyl phenol or tribromophenol, a long-chain alkylphenol, aliphatic carboxylic acid chloride, aliphatic carboxylic acid, hydroxy-benzoic acid alkyl ester, alkylether-phenol or the like. In the preparation of the polycarbonate resin used in the present invention, these terminators or molecular weight modifiers can be used singly or in the form of the mixture thereof.

The polycarbonate resin used in the present invention has a viscosity-average molecular weight of 10,000 to 100,000, preferably 15,000 to 50,000 calculated from the solution viscosity measured at 25°C in terms of a solution viscosity using methylene chloride as a solvent.

The oxy group- or a carbonyl group-containing aromatic compounds used in the composition according to the present invention, are represented by the following general formula (I) or (II):

$$(R^1)_h \qquad (X^1-R^2)_i$$

$$(R^3)_j \longrightarrow X^2 - R^4 - X^3 \longrightarrow (R^5)_k$$

where R^1 , R^3 and R^5 are independently a $(C_1 - C_{10})$ alkyl group, a $(C_1 - C_{10})$ alkoxy group, a $(C_1 - C_{10})$ alkoxy $(C_1 - C_{10})$ alkoxy $(C_1 - C_{10})$ alkyl group, -CH₂OH or a halogen atom; R^4 is a $(C_1 - C_{15})$ alkylene group, a $(C_2 - C_{15})$ alkylene group, a $(C_6 - C_{30})$ arylene group, a $(C_1 - C_{15})$ alkylene-dioxy group, or a $(C_6 - C_{30})$ arylene-di($C_1 - C_{15}$)alkylene-oxy group in which the said arylene group or the said arylene-dialkylene-oxy group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a $(C_1 - C_{10})$ alkyl group or a halogen atom; independently a hydrogen atom, a $(C_1 - C_{30})$ alkyl group, a $(C_3 - C_{30})$ -co- $(C_3 - C_{30})$ -alkoxy group, a $(C_6 - C_{30})$ -alkoxy group, the said arylalkoxy group, the said arylalkyl group, the said arylalkyl group, the said arylalkyl group, the said arylalkyl group or a halogen atom; group(s) bonded to the aromatic ring thereof, the substituent group(s) being a $(C_1 - C_{10})$ -alkyl group or a halogen atom; the range of 1 to 6, and if i is an integer not less than 2, the R^2 -s contained in the substituent groups bonded to the same aromatic ring may be covalently bonded with each other.

The aromatic compound containing oxy group or carbonyl groups may be used singly or in the form of the mixture thereof.

Specific examples of the aromatic compound containing oxy group or carbonyl groups represented by the general formula (i) may include diphenyl ether, arylphenyl ether, dibenzyl ether, benzylmethyl ether, benzylphenyl ketone, b

Specific examples of the aromatic compound containing oxy group or carbonyl groups represented by the general formula (II) may include dibenzoyl methane, dibenzoyl propane, 1, 2-dibenzyloxy ethane, hydroquinone-dibenzyl ether, ethylene glycol dibenzoate, or the like.

The blending amount of the aromatic compound containing oxy group or carbonyl group is in the range of 0.01 to 5 parts by weight based on 100 parts by weight of the polycarbonate resin. If the blending amount of the aromatic compound containing oxy group or carbonyl group is less than 0.01 part by weight, the aimed effect of preventing yellow discoloration upon exposure to an ionizing radiation cannot be sufficiently achieved. On the other hand, if the blending amount of the aromatic compound containing oxy group or carbonyl group is more than 5 parts by weight, mechanical properties of the resultant polycarbonate resin composition are deteriorated. In order to attain both the effect of prevent-blending amount of the aromatic compound containing oxy group or carbonyl group is preferably in the range of 0.05 to 3 parts by weight based on 100 parts by weight of the polycarbonate resin.

(A): The compound containing cyclic acetal groups used in the composition according to the present invention, are represented by the following formula (III) or (IV):

$$(R^{9}R^{10}C)_{p}$$
 $HC - (X^{4} + R^{11}(X^{5})_{m} CH)_{0} (CR^{12}R^{13})_{q}$ (III)

10

15

$$(R^{14}R^{15}C)_{r} \xrightarrow{HC} (X^{6} \xrightarrow{n} R^{16})$$
(IV)

where R^9 , R^{10} , R^{12} , R^{13} , R^{14} and R^{15} are independently a hydrogen atom, a $(C_1\text{-}C_{10})$ alkyl group, - CH_2OH , - $COOCH_3$, group, in which the said arylene group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a $(C_1\text{-}C_{10})$ alkyl group or a halogen atom; R^{16} is a hydrogen atom, a $(C_1\text{-}C_{30})$ alkyl group, a $(C_2\text{-}C_{30})$ alkenyl group, a $(C_2\text{-}C_{30})$ alkenyl group, a $(C_6\text{-}C_{30})$ aryl group, a $(C_6\text{-}C_{30})$ alkyl group, a $(C_6\text{-}C_{30})$ aryl $(C_1\text{-}C_{30})$ alkyl group, a $(C_6\text{-}C_{30})$ aryl group, the said arylalkyl group, the said arylalkyl group, the said arylalkyl group or the said arylacyl group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a $(C_1\text{-}C_{10})$ alkyl group or a halogen atom; $(C_6\text{-}C_{30})$ aryl group; and I, m and n are independently an integer of 0 or 1; and p, q and r are independently an integer of 1 to 10, and if p, q or r is not less than 2, the two or more of each of R^9 , R^{10} , R^{12} and R^{13} in the general formula (IV) may be the same or different.

These compound containing cyclic acetal groups may be used singly or in the form of the mixture thereof. Specific examples of the compound containing cyclic acetal groups represented by the general formula (III) may include 2, 2'-ethylene-bis-1, 3-dioxolane, 2, 2'-trimethylene-bis-1, 3-dioxolane, 2, 2'-phenylene-bis-1, 3-dioxolane, or the

Specific examples of the compound containing cyclic acetal groups represented by the general formula (IV) may include 1,3-dioxolane, 2-methyl-1, 3-dioxolane, 2-methyl-4-methyl-1, 3-dioxolane, 2-pentyl-4-methyl-1, 3-dioxolane, 2-pentyl-4-methyl-1, 3-dioxolane, 2-bromomethyl-1, 3-dioxolane, 2-chloromethyl-1, 3-dioxolane, 2-formomethyl-1, 3-dioxolane, 2-formomethyl-1

The blending amount of the compound containing cyclic acetal group which is represented by the general formula (III) or (IV), is in the range of 0.01 to 5 parts by weight based on 100 parts by weight of the polycarbonate resin. If the blending amount of the compound containing cyclic acetal group is less than 0.01 parts by weight, the aimed effect of preventing the yellow discoloration upon exposure to an ionizing radiation cannot be sufficiently achieved. On the other hand, if the blending amount of the compound containing cyclic acetal group is more than 5 parts by weight, mechanical properties of the resultant polycarbonate resin composition are deteriorated. In order to attain both the effect of preventing the yellow discoloration and the deterioration of the mechanical properties under well-balanced conditions, the blending amount of the compound containing cyclic acetal group is preferably in the range of 0.05 to 3 parts by weight

The weight ratio of the compound containing cyclic acetal group represented by the general formula (III) or (IV) to the aromatic compound containing oxy group or carbonyl group represented by the general formula (I) or (II) is not particularly limited, but is preferably in the range of 10/90 to 90/10, more preferably 20/80 to 80/20.

As described above, when the aromatic compound containing oxy group or carbonyl group and the compound containing cyclic acetal group represented by the general formula (III) or (IV) are blended at specified blending ratios with properties and a very low yellow discoloration upon exposure to an ionizing radiation for sterilization thereof.

(B): The sulfone compounds used in the composition according to the present invention, are represented by the following formula (V), (VI) or (VII):

$$R^{18} - S - R^{19}$$
 (V)

15

20

55

$$\begin{array}{cccc}
O & R^{23} \\
O & R^{24}
\end{array}$$
(VII)

where R^{18} , R^{19} , R^{20} and R^{22} are independently a $(C_1 - C_{30})$ alkyl group, a $(C_3 - C_{30})$ cycloalkyl group, a $(C_2 - C_{30})$ alkenyl group, a (C_6-C_{30}) aryl group, a (C_6-C_{30}) aryl (C_1-C_{30}) alkyl group, a (C_6-C_{30}) aryl (C_2-C_{30}) alkenyl group, a (C_1-C_{30}) aryl (C_2-C_{30}) aryl (C_2-C_{30}) alkenyl group, a (C_1-C_{30}) aryl (C_2-C_{30}) aryl $(C_2-C_{30$ C_{30})acyl(C_1 - C_{30})alkyl group, a (C_1 - C_{30})alkoxy(C_1 - C_{30})alkyl group, a (C_6 - C_{30})aryl(C_1 - C_{30})alkoxy(C_1 - C_{30})alkyl group, or a (C_6-C_{30}) aryl (C_1-C_{30}) alkoxy (C_2-C_{30}) alkenyl group, in which the said aryl group, the said arylalkyl group, the said arylalkenyl group, the said arylalkoxyalkyl group and the said arylalkoxyalkenyl group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C₁-C₄)alkyl group, a halogen atom, -NO₂, - $N(R^{25})_2$. -OH, -CH₂OR²⁶ or -OCH₃ where R^{25} and R^{26} are independently a hydrogen atom, a (C₁-C₁₀)alkyl group, a (C_6-C_{30}) aryl group or a (C_6-C_{30}) aryl (C_1-C_{10}) alkyl group, and in which the said acylalkyl group, the said alkoxyalkyl group, the said arylalkoxyalkyl group and the said arylalkoxyalkenyl group may have substituent group(s) bonded to its alkyl or alkenyl chain and selected from a (C_1-C_{30}) acyl group, a (C_1-C_{30}) alkoxy group or a (C_6-C_{30}) aryl (C_1-C_{30}) alkoxy group, and R^{18} and R^{29} , or R^{20} and R^{22} may be covalently bonded with each other; R^{21} , R^{23} and R^{24} are independently a (C_1-C_{15}) alkylene group, a (C_2-C_{15}) alkenylene group, or a (C_6-C_{30}) arylene group, in which the said arylene group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C₁-C₄)alkyl group, a halogen atom, -NO₂, -N(R²⁵)₂, -OH, -CH₂OR²⁶ or -OCH₃ where R²⁵ and R²⁶ have the same meaning as defined

These sulfone compounds may be used singly or in the form of the mixture thereof.

Specific examples of the sulfone compounds represented by the general formula (V) may include dimethyl sulfone, dibutyl sulfone, diphenyl sulfone, dihydroxyphenyl sulfone, ditolyl sulfone, dixylyl sulfone, dimesityl sulfone, dinitrophenyl sulfone, diaminophenyl sulfone, dichlorophenyl sulfone, dimethoxyphenyl sulfone, divinyl sulfone, dicyclohexyl sulfone, dibenzyl sulfone, 1-propenylmethyl sulfone, methylphenyl sulfone, methyltolyl sulfone, methylcyclohexyl sulfone, methylmethoxyphenyl sulfone, methylchlorophenyl sulfone, methylallyl sulfone, methylbenzyl sulfone, phenylallyl sulfone, phenyltalyl sulfone, phenylxylyl sulfone, phenylmethoxyphenyl sulfone, phenylvinyl sulfone, phenylbenzyl sulfone, phenylstyryl sulfone, phenylcinnamyl sulfone, trimethyl-3-(phenylsulfonyl)ortho-propionate, phenylmethoxymethyl sulfone, phenylphenoxymethyl sulfone, phenylacetonyl sulfone, phenylphenathyl sulfone, phenylcyclohexyl sulfone, phenylmethyl-benzene-sulfone, phenylmethoxycarbonyl sulfone, tolylallyl sulfone, tolylbutyl sulfone, tolylphenathyl sulfone, tolylbenzyl sulfone, tolylaminophenyl sulfone, tolylchlorobenzyl sulfone, 4-(toluene-sulfonyl)butane-2-on, benzylallyl sulfone, trimethylene sulfone, tetramethylene sulfone, pentamethylene sulfone, benzothiophene dioxide, dibenzothiophene dioxide, thioxanthene-9, 9-dioxide, 2, 5-dihydroxythiophene-1, 1-dioxide, 4-butylthian-1,1-dioxide, or the like.

Specific examples of the sulfone compounds represented by the general formula (VI) may include bis(methylsulfonyl)methane, 2, 2-bis(methylsulfonyl)propane, bis(phenylsulfonyl)methane, 1, 2-bis(phenylsulfonyl)ethane, 1, 2bis(phenylsulfonyl)ethylene, phenylsulfonyl-benzylsulfonyl-phenyl methane, or the like.

Specific examples of the sulfone compounds represented by the general formula (VII) may include 1, 4-thioxane-1, 1-dioxide, or the like.

The blending amount of the sulfone compound, which is represented by the general formula (V). (VI) or (VII), is in the range of 0.01 to 5 parts by weight based on 100 parts by weight of the polycarbonate resin. If the blending amount of the sulfione compound is less than 0.01 parts by weight, the aimed effect of preventing the yellow discoloration upon application of an ionizing radiation cannot be sufficiently achieved. On the other hand, if the blending amount of the sultone compound is more than 5 parts by weight, mechanical properties of the resultant polycarbonate resin composition

are unsuitably deteriorated. In order to attain both the effect of preventing the yellow discoloration and the deterioration of the mechanical properties under well-balanced conditions, the blending amount of the sulfone compound is preferably in the range of 0.05 to 3 parts by weight based on 100 parts by weight of the polycarbonate resin.

The weight ratio of the sulfone compound represented by the general formula (V). (VI) or (VII) to the aromatic compound containing oxy group or carbonyl group represented by the general formula (I) or (II) is not particularly limited.

As described above, when the aromatic compound containing oxy group or carbonyl group and the sulfone compound represented by the general formula (V), (VI) or (VII) are blended at specified blending ratios with the polycarbonate resin, the resultant polycarbonate resin composition can exhibit no deterioration of its mechanical properties and a very low yellow discoloration upon exposure to an ionizing radiation for sterifization thereof.

(C): The sulfoxide compounds used in the composition according to the present invention, are represented by the following formula (VIII), (IX) or (X):

15

20

25

30

$$R^{27} = S - R^{28}$$
O
(VIII)
$$R^{29} = S - (R^{30} - S) - R^{31}$$
O
O
(IX)

where R^{27} , R^{28} , R^{29} , R^{31} , R^{32} and R^{35} are independently a (C_1-C_{30}) alkyl group, a (C_3-C_{30}) cycloalkyl group, a (C_2-C_{30}) C_{30})alkenyl group, a (C_6 - C_{30})aryl group, a (C_6 - C_{30})aryl(C_1 - C_{30})alkyl group, a (C_6 - C_{30})aryl(C_2 - C_{30})alkenyl group, a (C_1 - C_2) C₃₀)acyl(C₁-C₃₀)alkyl group, a (C₁-C₃₀)alkoxy(C₁-C₃₀)alkyl group, a (C₆-C₃₀)aryl(C₁-C₃₀)alkoxy(C₁-C₃₀)alkyl group, a (C₆-C₃₀)aryl(C₁-C₃₀)alkoxy(C₂-C₃₀)alkenyl group, a (C₁-C₃₀)alkoxy group, a (C₁-C₃₀)alkoxycarbonyl group, a polyethylene-glycol-ether group or a pyridyl group, in which the said aryl group, the said arylalkyl group, the said arylalkenyl group, the said arylalkoxyalkenyl group and the said arylalkoxyalkyl group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C₁-C₄)alkyl group, a halogen atom, -NO₂, -N(R³⁶)₂, -COOH, -COOCH₃, -OH, -CH₂OR³⁷ or -OCH₃, and in which the said acylalkyl group, the said alkoxyalkyl group, the said aryla-Ikoxyalkenyl group and the said arylalkoxyalkyl group may have substituent group(s) bonded to the alkyl or alkenyl chain thereof, the substituent group(s) being a (C_1-C_{30}) acyl group, a (C_1-C_{30}) alkoxy group or a (C_6-C_{30}) aryl (C_1-C_{30}) alkoxy group; R^{27} and R^{28} , R^{29} and R^{31} or R^{32} and R^{35} may be covalently bonded with each other; R^{30} , R^{33} and R^{34} are independently a (C₁-C₁₅)alkylene group, a (C₂-C₁₅)alkenylene group, a (C₆-C₃₀)arylene group, a (C₆-C₃₀)arylenedi(C₁-C₁₅)alkylene group, or -O(CH₂CHR³⁸O)_t-, in which the said arylene group and the said arylenedialkylene group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C₁-C₄)alkyl group, a halogen atom, -NO₂, -N(R³⁶)₂, -COOH, -COOCH₃, -OH, -CH₂OR³⁷ or -OCH₃, and R³³ and R³⁴ are different from each other; R³⁶, R³⁷ and R³⁸ are independently a hydrogen atom, a (C₁-C₁₀)alkyl group, a (C₆-C₁₅)aryl group or a (C₆-C₁₅-C₁₅-C₁₅-C₁₅-C₁₅-C₁₅-C₁₅-C₁₅-C₁₅-C₁₅-C₁₅-C₁₅ C₁₅)aryl(C₁-C₁₀)alkyl group; and b, c and t are independently an integer of 1 to 100.

These sulfoxide compounds may be used singly or in the form of the mixture thereof.

Specific examples of the sulfoxide compounds represented by the general formula (VIII) may include dimethyl sulfaxide, dibutyl sulfoxide, diphenyl sulfoxide, dihydroxyphenyl sulfoxide, ditolyl sulfoxide, dibenzyl sulfoxide, divinyl sulfoxide, dixylyl sulfoxide, dimesityl sulfoxide, dinitrophenyl sulfoxide, diaminophenyl sulfoxide, dichlorophenyl sulfoxide, dimethoxyphenyl sulfoxide, dicyclohexyl sulfoxide, 1-propenylmethyl sulfoxide, methylcyclohexyl sulfoxide, methylphenyl sulfoxide, methyltolyl sulfoxide, methylmethoxyphenyl sulfoxide, methylchlorophenyl sulfoxide, methylcarboxyphenyl sulfoxide, methylallyl sulfoxide, methylbenzyl sulfoxide, phenylallyl sulfoxide, phenyltolyl sulfoxide, phenylxylyl sulfoxide, phenylmethoxyphenyl sulfoxide, phenylvinyl sulfoxide, phenylbenzyl sulfoxide, phenylstyryl sulfoxide, phenylcinnamyl sufficially, suffi ide, phenylacetonyl sulfoxide, phenylphenathyl sulfoxide, phenylcyclohexyl sulfoxide, phenylmethylbenzyl sulfoxide, phenylmethoxycarbonyl sulfoxide, tolylbenzyl sulfoxide, tolylbutyl sulfoxide, tolylbenathyl sulfoxide, tolylbutoxy sulfox-

ide, tolylallyl sulfoxide, tolylaminophenyl sulfoxide, tolylchlorobenzyl sulfoxide, 4-(toluene-sulfinyl)butane-2-on, benzylaltyl sulfoxide, 2-pyridylbenzyl sulfoxide, acetonylnitrophenyl sulfoxide, trimethylene sulfoxide, tetramethylene sulfoxide, pentamethylene sulfoxide, benzothiophene-1-oxide, dibenzothiophene-1-oxide, thioxanthene-9-oxide, 2, 5-dihydroxythiophene-1-oxide, 4-butylthian-1-oxide, or the like.

Specific examples of the sulfoxide compounds represented by the general formula (IX) may include bis(methytsulfinyl)methane, 2, 2-bis(methylsulfinyl)propane, bis(phenylsulfinyl)methane, 1, 2-bis(phenylsulfinyl)ethane, 1, 2-bis(phenylsulfinyl)ethane, 1, 2-bis(phenylsulfinyl)ethane, 2, 2-bis(methylsulfinyl)ethane, 3, 2-bis(phenylsulfinyl)ethane, 3, 3-bis(phenylsulfinyl)ethane, 3, 3-bis(phenylsulfi nytsulfinyl)ethylene, phenylsulfinyl-benzylsulfinylphenyl methane, 1,4-dithian-1, 4-dioxide, polytetramethylene suffoxide, polyhexamethylene suffoxide, or the like.

Specific examples of the sulfoxide compounds represented by the general formula (X) may include poly(hexamethy l enesul foxide-co-octamethylenes ulfoxide). poly (phenylenes ulfoxide-co-hexamethylenes ulfoxide). poly (phenylenes ulfoxide-co-hexamethylenes ulfoxide).nesulfoxide-co-3, 6, 9, 12, 15, 18-hexaoxyeicosanylene-sulfoxide), or the like.

The blending amount of the sulfoxide compound, which is represented by the general formula (VIII), (IX) or (X), is in the range of 0.01 to 5 parts by weight based on 100 parts by weight of the polycarbonate resin. If the blending amount of the said sulfoxide compound is less than 0.01 parts by weight, the aimed effect of preventing the yellow discoloration upon exposure to an ionizing radiation cannot be sufficiently achieved. On the other hand, if the blending amount of the said sulfoxide compound is more than 5 parts by weight, mechanical properties of the resultant polycarbonate resin composition are unsuitably deteriorated. In order to attain both the aimed effect of preventing the yellow discoloration and the deterioration of the mechanical properties under well-balanced conditions, the blending amount of the sufficide compound is preferably in the range of 0.05 to 3 parts by weight based on 100 parts by weight of the polycarbonate

The weight ratio of the sulfoxide compound represented by the general formula (VIII), (IX) or (X) to the aromatic compound containing oxy group or carbonyl group represented by the general formula (I) or (II) is not particularly limited, but is preferably in the range of 10/90 to 90/10, more preferably 20/80 to 80/20.

As described above, when the aromatic compound containing oxy group or carbonyl group and the sulfoxide compound represented by the general formula (VIII), (IX) or (X) are blended at specified blending ratios with the polycarbonate resin, the resultant polycarbonate resin composition can exhibit no deterioration of its mechanical properties and a very low yellow discoloration upon exposure to an ionizing radiation for sterilization thereof.

(D) The polyalkylene glycol, the ether of polyalkylene glycol or the ester of polyalkylene glycol (hereinafter referred to as polyalkylene glycol derivative) used in the composition according to the present invention may be those represented by the general formula (XI) or (XII):

$$R^{39}O - \{ (CH_2CH) - O \}_e - R^{41}$$
 (XI)

$$R^{42}COO - \{ (CH_2CH) - O \}_g - COR^{44}$$
 (XII)

35

where R³⁹, R⁴¹, R⁴² and R⁴⁴ are independently a hydrogen atom, a (C₁-C₃₀)alkyl group, a (C₃-C₃₀)cycloalkyl group, a (C_2-C_{30}) alkenyl group, a (C_6-C_{30}) aryl group, a (C_6-C_{30}) aryl (C_1-C_{30}) alkyl group or a (C_6-C_{30}) aryl (C_2-C_{30}) alkenyl group, in which the said aryl group, the said arylalkyl group and the said arylalkenyl group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C₁-C₁₀)alkyl group or a halogen atom; R⁴⁰ and R⁴³ are independently a hydrogen atom or a (C₁-C₄)alkyl group; e and g are independently an integer of not less than 1, preferably 1 to 1000, and d and f are independently an integer of 1 to 10.

These polyalkylene glycol derivatives may be used singly or in the form of the mixture thereof.

Specific examples of the polyalkylene glycol derivatives represented by the general formula (XI) may include polyethylene glycol, polyethylene glycol methylether, polyethylene glycol dimethylether, polyethylene glycol dodecylether, polyethylene glycol benzylether, polyethylene glycol dibenzylether, polyethylene glycol-4-nonylphenylether, polypropylene glycol, polypropylene glycol methylether, polypropylene glycol dimethylether, polypropylene glycol dodecylether, polypropylene glycol benzylether, polypropylene glycol dibenzylether, polypropylene glycol-4-nonylphenylether, polyte-

Specific examples of the polyalkylene glycol derivatives represented by the general formula (XII) may include pol-

yethylene glycol diacetate, polyethylene glycol-(monoacetate)monopropionate, polyethylene glycol dibutyrate, polyethylene glycol distearate, polyethylene glycol dibenzoate, polyethylene glycol di-2, 6-dimethyl-benzoate, polyethylene głycol di-p-tert-butyl-benzoate, polyethylene glycol dicaprylate, polypropylene glycol diacetate, polypropylene głycol-(monoacetate)monopropionate, polypropylene glycol dibutyrate, polypropylene glycol distearate, polypropylene glycol dibenzoate, polypropylene glycol di-2, 6-dimethyl-benzoate, polypropylene glycol di-p-tert-butyl-benzoate, polypropyl-

The blending amount of the polyalkylene glycol derivative, which is represented by the general formula (XI) or (XII). is in the range of 0.01 to 5 parts by weight based on 100 parts by weight of the polycarbonate resin. If the blending amount of the compound is less than 0.01 parts by weight, the aimed effect of preventing the yellow discoloration upon exposure to an ionizing radiation cannot be sufficiently achieved. On the other hand, if the blending amount of the polyalkylene glycol derivative is more than 5 parts by weight, mechanical properties of the resultant polycarbonate resin composition are unsuitably deteriorated. In order to attain both the aimed effect of preventing the yellow discoloration and the deterioration of the mechanical properties under well-balanced conditions, the blending amount of the polyalkylene glycol derivative is preferably in the range of 0.05 to 3 parts by weight based on 100 parts by weight of the polycarbonate resin.

The weight ratio of the polyalkylene glycol derivative represented by the general formula (XI) or (XII) to the aromatic compound containing oxy group or carbonyl group represented by the general formula (I) or (II) is not particularly limited, but is preferably in the range of 10/90 to 90/10, more preferably 20/80 to 80/20.

As described above, when the aromatic compound containing oxy group or carbonyl group and the afore-mentioned polyalkylene glycol derivative of the general formula (XI) or (XII) are blended at specified blending ratios with the polycarbonate resin, the resultant polycarbonate resin composition shows no deterioration of its mechanical properties and a very low yellow discoloration when exposed to an ionizing radiation for sterilization thereof.

(E): The compound containing sulfonate groups used in the composition according to the present invention, are represented by the following formula (XIII), (XIV) or (XV):

25

35

$$(R^{45})_{s} = \begin{pmatrix} 0 \\ | \\ | \\ | \\ 0 \end{pmatrix}$$
 (XIII)

$$(R_{50})_{x} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}_{y} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}_{z}$$
(XV)

where R^{45} , R^{47} , R^{50} and R^{52} are independently a (C_1-C_{30}) alkyl group, a (C_6-C_{30}) arylsulfone group or a halogen atom; R⁴⁶ and R⁴⁹ are independently a hydrogen atom, a (C₁-C₃₀)alkyl group, a (C₃-C₃₀)cycloalkyl **group**, a (C_2-C_{30}) alkenyl group, a (C_6-C_{30}) aryl group, a (C_6-C_{30}) aryl (C_1-C_{30}) alkyl group, a (C_6-C_{30}) aryl (C_2-C_{30}) alkenyl group or a (C1-C30) acyl group, in which the said aryl group, the said arylalkyl group and the said arylalkenyl group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C1-C4)alkyl group or a halogen atom; R^{48} and R^{51} are independently a (C_1-C_{15}) alkylene group, a (C_2-C_{15}) alkenylene group or a (C_6-C_{15}) alkylene group C₃₀)arylene group, in which the said arylene group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C_1-C_4) alkyl group or a halogen atom; s, v, x and z are independently an integer of 0 to 5 and u is an integer of 1 to 6, preferably 1 to 3 with provided that a sum of s and u is in the range of 1 to 6; and w

and y are independently an integer of not less than 1, preferably from 1 to 500.

These compound containing sulfonate groups may be used singly or in the form of the mixture thereof.

Specific examples of the compound containing sulfonate groups represented by the general formula (XIII) may include methyl benzene-sulfonate, phenyl benzene-sulfonate, methyl toluene-sulfonate, ethyl toluene-sulfonate, butyl toluene-sulfonate, phenyl toluene-sulfonate, phenyl toluene-sulfonate, phenyl toluene-sulfonate, phenyl toluene-sulfonate, methyl toluene-sulfonate, naphthyl toluene-sulfonate, methyl mesitylene-sulfonate, phenyl toluene-sulfonate, methyl mesitylene-sulfonate, phenyl mesitylene-sulfonate, phenyl benzene-disulfonate, diphenyl-sulfone-3-methyl sulfonate, diphenyl-sulfone-3-phenyl sulfonate, or the like.

Specific examples of the compound containing sulfonate groups represented by the general formula (XIV) may include methoxyethyl benzene-sulfonate, methoxymethyl toluene-sulfonate, methoxyethyl toluene-sulfonate, ethoxyethyl toluene-sulfonate, benzyloxyethyl toluene-sulfonate, methoxyethyl toluene-sulfonate, benzyloxyethyl toluene-sulfonate, methoxyethyl mesitylene-sulfonate, butoxyethoxyethyl toluene-sulfonate, phenoxyethoxyethyl toluene-sulfonate, benzyloxyethoxyethyl toluene-sulfonate, or the like.

Specific examples of the compound containing sulfonate groups represented by the general formula (XV) may include bis-tosyloxy methane, 1, 2-bis-tosyloxy ethane, diethylene glycol-di-p-tosylate, tetraethylene glycol-di-p-tosylate, or the like.

The blending amount of the compound containing sulfonate group, which is represented by the general formula (XIII), (XIV) or (XV), is in the range of 0.01 to 5 parts by weight based on 100 parts by weight of the polycarbonate resin. If the blending amount of the compound containing sulfonate group is less than 0.01 parts by weight, the aimed effect of preventing the yellow discoloration upon exposure to an ionizing radiation cannot be sufficiently achieved. On the other hand, if the blending amount of the compound containing sulfonate group is more than 5 parts by weight, mechanical properties of the resultant polycarbonate resin composition are unsuitably deteriorated. In order to attain both the aimed effect of preventing the yellow discoloration and the deterioration of the mechanical properties under well-balanced conditions, the blending amount of the compound containing sulfonate group is preferably in the range of 0.05 to 3 parts by weight based on 100 parts by weight of the polycarbonate resin.

The weight ratio of the compound containing sulfonate group represented by the general formula (XIII). (XIV) or (XV) to the aromatic compound containing oxy group or carbonyl group represented by the general formula (I) or (II) is not particularly limited, but is preferably in the range of 10/90 to 90/10, more preferably 20/80 to 80/20.

As described above, when the aromatic compound containing oxy group or carbonyl group and the compound containing sulfonate group represented by the general formula (XIII), (XIV) or (XV) are blended at specified blending ratios with the polycarbonate resin, the resultant polycarbonate resin composition can exhibit no deterioration of its mechanical properties and a very low yellow discoloration upon exposure to an ionizing radiation for sterilization thereof.

(F). The aromatic hydrocarbon-aldehyde resins used in the composition according to the present invention may be prepared by reacting aromatic hydrocarbon with aldehyde in the presence of an acid catalyst.

Specific examples of the aromatic hydrocarbons used for the preparation of the aromatic hydrocarbon-aldehyde resins may include monocyclic aromatic hydrocarbon compounds such as benzene, toluene, ethyl benzene, xylene, methylethyl benzene, trimethyl benzene, tetramethyl benzene, pseudo-cumene or cumene, polycyclic aromatic hydrocarbon compounds such as naphthalene, methyl naphthalene, ethyl naphthalene, dimethyl naphthalene, acenaphthene, anthracene or the like. These aromatic hydrocarbon compounds may be used singly or in the form of the mixture thereof. Among them, the especially preferred aromatic hydrocarbon compounds are toluene, xylene, mesitylene, pseudo-cumene, naphthalene or the like.

Specific examples of the aldehydes used for the preparation of the aromatic hydrocarbon-aldehyde resins may include saturated aliphatic aldehydes such as formaldehyde, acetaldehyde, propionaldehyde, butylaldehyde, iso-butylaldehyde, valeraldehyde, laurinaldehyde or stearinaldehyde; aliphatic polyvalent aldehydes such as glyoxal or succindialdehyde; unsaturated aliphatic aldehydes such as acrolein, crotonaldehyde or propiolaldehyde; aromatic aldehydes such as benzaldehyde, tolylaldehyde, salicylaldehyde, cinnamaldehyde or naphthaldehyde; heterocydic aldehydes such as furfural; aldehyde derivatives such as methylal, dioxolane, trioxane, tetraoxane, paraformaldehyde, paraldehyde or metaldehyde, or the like.

These aldehydes may be used singly or in the form of the mixture thereof. Among them, the especially preferred aldehydes are formaldehyde, trioxane, paraformaldehyde, acetaldehyde, or the like.

It is preferred that the aromatic hydrocarbon-aldehyde resins contains substantially no acetal group and has such a structure that adjacent aromatic rings are mainly bonded through an alkylene group or alkylene-ether group with each other. The expression "substantially no acetal group" means that the acetal group is contained in an amount of not more than 0.1 mole based on one molecule of the aromatic hydrocarbon-aldehyde resins.

Such aromatic hydrocarbon-aldehyde resins containing substantially no acetal group are commercially available. For example, as the commercially available aromatic hydrocarbon-aldehyde resins, NICANOL DS, NICANOL S or NICANOL K (produced by Mitsubishi Gas Chemical Co., Ltd.), may be exemplified. Further, the aromatic hydrocarbon-aldehyde resins can be prepared according to methods disclosed in Japanese Patent Application Laid-open (Kokai) Nos. 60-51133 (1985), 61-23016 (1986), 61-213216 (1986), 63-196616 (1988), 4-224825 (1992), 4-335014 (1992), 5-

186544 (1993), 6-136081 (1994) and the like.

15

25

It is further preferred that the aromatic hydrocarbon-aldehyde resins used in the present invention, have an oxygen content of not less than 8 % by weight, preferably 9 to 25 % by weight. Such aromatic hydrocarbon-aldehyde resins are commercially available. For example, as the commercially available aromatic hydrocarbon-aldehyde resins having an oxygen content of not less than 8 % by weight, NICANOL H, NICANOL L, NICANOL G or NICANOL Y (produced by Mitsubishi Gas Chemical Co., Ltd.), GENERITE 6010 or GENERITE 5100 (produced by General Petroleum Chemical

These aromatic hydrocarbon-aldehyde resins may be used singly or in the from of the mixture thereof.

The blending amount of the aromatic hydrocarbon-aldehyde resins is in the range of 0.01 to 5 parts by weight, preferably 0.1 to 2 parts by weight based on 100 parts by weight of the polycarbonate resin. If the blending amount of the aromatic hydrocarbon-aldehyde resins is less than 0.01 parts by weight, the aimed effect of preventing the yellow discoloration upon exposure to an ionizing radiation cannot be sufficiently achieved. On the other hand, if the blending amount of the aromatic hydrocarbon-aldehyde resins is more than 5 parts by weight, mechanical properties, heat resistance, etc. of the resultant polycarbonate resin composition are unsuitably deteriorated.

The weight ratio of the aromatic hydrocarbon-aldehyde resins to the aromatic compound containing oxy group or carbonyl group represented by the general formula (I) or (II) is not particularly limited, but is preferably in the range of

As described above, when the aromatic compound containing oxy group or carbonyl group and the aromatic hydrocarbon-aldehyde resins are blended at specified blending ratios with the polycarbonate resin, the resultant polycarbonate resin composition can exhibit no deterioration of its mechanical properties and a very low yellow discoloration upon exposure to an ionizing radiation for sterilization thereof.

Further, among the polycarbonate resin compositions comprising the polycarbonate resin, the aromatic compound containing oxy group or carbonyl group and at least two compound selected from the group consisting of the afore-mentioned compounds (A) to (F), the following polycarbonate resin composition is preferred.

The polycarbonate resin composition contains 100 parts by weight of the polycarbonate resin, 0.01 to 5 parts by weight of the aromatic compound containing oxy group or carbonyl group, 0.01 to 5 parts by weight of the compound containing cyclic acetal group of the general formula (III) or (IV) and 0.01 to 5 parts by weight of the polyalkylene glycol, the ether of polyalkylene glycol or the ester of polyalkylene glycol of the general formula (XI) or (XII).

In this case, as described previously, the blending amount of the compound containing cyclic acetal group represented by the general formula (III) or (IV) is in the range of 0.01 to 5 parts by weight based on 100 parts by weight of the polycarbonate resin. If the blending amount of the compound containing cyclic acetal group is less than 0.01 parts by weight, the aimed effect of preventing the yellow discoloration upon exposure to an ionizing radiation cannot be sufficiently achieved. On the other hand, if the blending amount of the said compound containing cyclic acetal group is more than 5 parts by weight, mechanical properties of the resultant polycarbonate resin composition are deteriorated. In order to attain both the aimed effect of preventing the yellow discoloration and the deterioration of the mechanical properties under well-balanced conditions, the blending amount of the said compound containing cyclic acetal group is preferably in the range of 0.05 to 3 parts by weight based on 100 parts by weight of the polycarbonate resin.

Further, in this case, as also described previously, the blending amount of the polyalkylene glycol derivative represented by the general formula (XI) or (XII) is in the range of 0.01 to 5 parts by weight based on 100 parts by weight of the polycarbonate resin. If the blending amount of the said polyalkylene glycol derivative is less than 0.01 parts by weight, the resultant polycarbonate resin composition cannot exhibit the aimed effect of preventing the yellow discoloration upon exposure to an ionizing radiation and mold release characteristics to a sufficient extent. On the other hand, if the blending amount of the said polyalkylene glycol derivative is more than 5 parts by weight, mechanical properties of the resultant polycarbonate resin composition are unsuitably deteriorated. In order to attain the aimed effect of preventing the yellow discoloration, and the deterioration of the mechanical properties and the mold-release properties under well-balanced conditions, the blending amount of the said polyalkylene glycol derivative is preferably in the range of 0.05 to 3 parts by weight based on 100 parts by weight of the polycarbonate resin.

In the afore-mentioned especially preferred polycarbonate resin composition, the blending percentage of each of the aromatic compound containing oxy group or carbonyl group, the compound containing cyclic acetal group and the polyalkylene glycol derivative, is preferably in the range of 5 to 90 % by weight, more preferably 15 to 80 % by weight based on the total weight of these three compounds, though not particularly limited thereto. Thus, by blending the aromatic compound containing oxy group or carbonyl group, the compound containing cyclic acetal group, and the polyalkylene glycol derivative at specified blending percentages with the polycarbonate resin, there can be produced a polycarbonate resin composition capable of exhibiting a very low yellow discoloration without deterioration of its mechanical properties and excellent mold-release properties.

In the present invention, the blending of the aromatic compound containing oxy group or carbonyl group and at least one compound selected from the group consisting of the afore-mentioned compounds (A) to (F) with the polycarbonate resin, can be carried out at any optional stage up to the production of a final molded product according to various methods known in the art. Examples of the blending methods may include a mixing method using a tumbler, a Henschel

mixer or the like, a mixing method in which the afore-mentioned components are quantitatively fed into a hopper of an extruder, or the like.

The polycarbonate resin composition according to the present invention may further contain other additives to impart appropriate properties thereto, if required. Examples of the additives may include flame-retardants such as halogen compounds, phosphorus compounds or metal salts of sulfonic acid; flame-retardant assistants such as antimony compounds or zirconium compounds; melt drop- preventing agents upon ignition such as polytetrafluoroethylene or silicon compounds; impact modifiers such as elastomers; anti-oxidants; heat stabilizers; ultraviolet light absorbers; antistatic agents; plasticizers; mold release agents; lubricants; compatibilizing agents; foaming agents; reinforcing agents such as glass fiber, glass beads, glass flakes, carbon fiber, fibrous magnesium, potassium titanate whiskers, ceramic whiskers, mica or talc; filler; pigments; or the like. These additives may be used singly or in combination.

The polycarbonate resin composition can be formed into a molded product according to a conventional molding method such as an injection-molding method, a blow-molding method or the like. Specific examples of the medical supplies and appliances to which the polycarbonate resin composition according to the present invention is suitably applied, include an artificial dialyzer, an artificial lung, an anesthetic inhaler, a vein connector or accessories, a hemocentrifugal bowl, surgical appliances, appliances for an operation room, tubes for feeding oxygen into blood, connectors for tubes, cardiac probes and injectors, containers for the surgical appliances, the appliances for an operation room or an intravenous injection liquid, or the like.

The polycarbonate resin composition according to the present invention can show a very low yellow discoloration when exposed to an ionizing radiation for sterilization while maintaining inherent properties of polycarbonate resin, and excellent mold-release properties. Accordingly, the polycarbonate resin composition according to the present invention is useful for medical products, medical apparatuses, components for such medical apparatuses, or the like.

EXAMPLES:

30

The present invention is described in more detail below by way of the examples. However, the examples are only illustrative and therefore the present invention is not limited to these examples.

Examples 1 to 6 and Comparative Examples 1 to 3:

An aromatic compound containing oxy group or carbonyl group and a compound containing cyclic acetal group shown in Table 1 were blended, at blending ratios also shown in Table 1, with 100 parts by weight of a polycarbonate resin (lupiron S-2000 having a viscosity-average molecular weight of 25,000, produced by Mitsubishi Engineering Plastics Co., Ltd.) in a tumbler. The mixture was fed into a vented single-screw extruder having an screw diameter of 40 mmØ and extruded into pellets at a barrel temperature of 270°C.

The thus-prepared pellets were dried in a hot-air drier at a temperature of 120°C for not less than 5 hours. Thereafter, the dried pellets were injection-molded at a resin temperature of 270°C and mold temperature of 80°C to prepare a test specimen having a diameter of 50 mm2 and a thickness of 3 mm.

The thus-obtained test specimens were exposed to 25 kGy of a cobalt-60 gamma radiation and then they were measured the change in yellowness index (ΔYI) thereof. The measurements of the yellowness index of the test specimens were carried out according to JIS K7103 by using a color difference meter (SM-3-CH, manufactured by SUGA Test Instruments Co., Ltd). The results are also shown in Table 1.

Meanwhile, the yellowness index of the polycarbonate resin composition according to the present invention was normally not more than 9.5.

45

Table 1

Example No.	Aromatic compound containing oxy group or carbonyl group/compound containing cyclic acetal group	Blending ratio (part by weight)	ΔΥΙ
Example 1	dibenzyl ketone/2,2'-trimethylene bis-1,3-dioxolane		-
Example 2	dibenzyl ether/2-benzyl-1,3-dioxolane	0.2/0.5	9.3
Example 3		0.2/0.2	8.7
Example 4	benzoin/2-benzyl-1,3-dioxolane	0.5/0.5	6.2
Example 5	1,2-dibenzyloxy ethane/2-methoxy-1,3-dioxolane	0.5/0.75	7.9
	benzyl benzoate/2-benzyl-4-methyl-1,3-dioxolane	1.0/0.25	8.2
Example 6	dicyclohexyl phthalate/4-phenyl-1,3-dioxane	0.5/0.5	
Comparative Example 1	Not added	0.3/0.5	7.2
Comparative Example 2	benzyl benzoate/-	•	18.1
Comparative Example 3		1.0/-	10.3
	-/2-benzyl-1,3-dioxolane	-/0.5	10.5

Examples 7 to 12 and Comparative Examples 4 to 5:

An aromatic compound containing oxy group or carbonyl group and a sulfone compound shown in Table 2 were blended, at blending ratios also shown in Table 2, with 100 parts by weight of a polycarbonate resin (lupiron S-2000 having a viscosity-average molecular weight of 25,000, produced by Mitsubishi Engineering Plastics Co., Ltd.) in a tumbellets at a barrel temperature of 270°C.

The thus-prepared pellets were dried in a hot-air drier at a temperature of 120°C for not less than 5 hours. Thereafter, the dried pellets were injection-molded at a resin temperature of 270°C and a mold temperature of 80°C to prepare a test specimen having a diameter of 50 mmØ and a thickness of 3 mm.

The thus-prepared test specimens were exposed to 25 kGy of a cobalt-60 gamma radiation and then they were measured the change in yellowness index (ΔΥΙ) thereof. The measurements of the yellowness index of the test specimens were carried out according to JIS K7103 by using a color difference meter (SM-3-CH, manufactured by SUGA Test Instruments Co., Ltd). The results are also shown in Table 2.

Table 2

	iable 2		
Example No.	Aromatic compound containing oxy group or carbonyl group/sulfone compound	Blending ratio (part by weight)	ΔΥ
Example 7	dibenzyl ketone/diphenyl sulfone		├
Example 8	dibenzyl ether/diphenyl sulfone	0.5/0.25	8.7
Example 9		0.5/0.5	6.9
	dibenzyl ether/dibenzyl sulfone	0.25/0.25	7.6
Example 10	benzyl benzoate/dibenzyl sulfone		
Example 11		0.75/0.25	9.5
Example 12	dicyclohexyl phthalate/dibenzyl sulfone	1.0/0.5	9.2
	dibenzyl ether/phenylphenathyl sulfone	0.25/0.25	7.9
Comparative Example 4	-/diphenyl sulfone		7.9
Comparative Example 5		-/1.5	13.1
	-/dibenzyl sulfone	-/0.5	11.6

50

10

20

Examples 13 to 15 and Comparative Example 6:

An aromatic compound containing oxy group or carbonyl group and a sulfoxide compound shown in Table 3 were blended, at blending ratios also shown in Table 3, with 100 parts by weight of a polycarbonate resin (lupiron S-2000 having a viscosity-average molecular weight of 25,000, produced by Mitsubishi Engineering Plastics Co., Ltd.) in a tumbler. The mixture was fed into a vented single-screw extruder having a screw diameter of 40 mm \oslash and extruded into pellets at a barrel temperature of 270°C.

The thus-prepared pellets were dried in a hot-air drier at a temperature of 120°C for not less than 5 hours. Thereafter, the dried pellets were injection-molded at a resin temperature of 270°C and a mold temperature of 80°C to prepare a test specimen having a diameter of 50 mmØ and a thickness of 3 mm.

The thus-prepared test specimens were exposed to 25 kGy of a cobalt-60 gamma radiation and then they were measured the change in yellowness index (ΔYI) thereof. The measurements of the yellowness index of the test specimens were carried out according to JIS K7103 by using a color difference meter (SM-3-CH, manufactured by SUGA Test Instruments Co., Ltd). The results are also shown in Table 3.

Table 3

Aromatic compound containing oxy group or carbonyl group/sulfoxide compound	Blending ratio (part by weight)	ΔΥΙ
dibenzyl ketone/phenylmethyl sulfoxide	0.5/0.5	9.1
dibenzyl ether/diphenyl sulfoxide	0.25/0.25	7.4
		8.2
-/diphenyl sulfoxide	<u> </u>	12.3
	dibenzyl ketone/phenylmethyl sulfoxide dibenzyl ether/diphenyl sulfoxide 1,2-dibenzyloxy ethane/phenylmethyl sulfoxide	or carbonyl group/sulfoxide compound (part by weight) dibenzyl ketone/phenylmethyl sulfoxide 0.5/0.5 dibenzyl ether/diphenyl sulfoxide 0.25/0.25 1,2-dibenzyloxy ethane/phenylmethyl sulfoxide 0.25/0.75

Examples 16 to 19 and Comparative Example 7:

An aromatic compound containing oxy group or carbonyl group and a polyalkylene glycol derivative shown in Table 4 were blended, at blending ratios also shown in Table 4, with 100 parts by weight of a polycarbonate resin in a tumbler. The mixture was fed into a vented single-screw extruder having a screw diameter of 40 mmØ and extruded into pellets at a barrel temperature of 270°C.

Specifically, the raw materials used in this example were as follows.

- (1) Polycarbonate resin:
- lupiron S-2000 having a viscosity-average molecular weight of 25,000, produced by Mitsubishi Engineering Plastics Co., Ltd.
- (2) Polyalkylene glycol derivative:
- PEG1000: polyethylene glycol having an average molecular weight of 1,000, or PPG2000: polypropylene glycol having a molecular weight of 2,000.

The pellets were then dried in a hot-air drier at a temperature of 120°C for not less than 5 hours. Thereafter, the dried pellets were injection-molded at a resin temperature of 270°C and a mold temperature of 80°C to prepare a test specimen having a diameter of 50 mmØ and a thickness of 3 mm.

The thus-prepared test specimens were exposed to 25 kGy of a cobalt-60 gamma radiation and then they were measured the change in yellowness index (ΔΥΙ) thereof. The measurements of the yellowness index of the test specimens were carried out according to JIS K7103 by using a color difference meter (SM-3-CH, manufactured by SUGA Test Instruments Co., Ltd). The results are also shown in Table 4.

25

40

20

Table 4

	Table 4		
Example No.	Aromatic compound containing oxy group or carbonyl group/polyalkylene glycol derivative	Blending ratio (part by weight)	ΔΥΙ
Example 16	1,4-bis-methoxymethyl benzene/PPG2000	0.5/0.5	-
Example 17	dibenzyl ether/PPG2000		6.9
Example 18	1,2-dibenzyłoxy ethane/PPG2000	0.5/0.5	8.2
Example 19	dicyclohexyl phthalate/PEG1000	0.5/0.5	9.2
Comparative Example 7	-/PPG2000	0.75/0.5	8.7
	02000	-/1.0	12.7

10

Examples 20 to 26 and Comparative Example 8:

An aromatic compound containing oxy group or carbonyl group and a compound containing sulfonate group shown in Table 5 were blended, at blending ratios also shown in Table 5, with 100 parts by weight of a polycarbonate resin (lupiron S-2000 having a viscosity-average molecular weight of 25,000, produced by Mitsubishi Engineering Plastics Co., Ltd.) in a tumbler. The mixture was fed into a vented single-screw extruder having a screw diameter of 40 mm/

The thus-prepared pellets were dried in a hot-air drier at a temperature of 120°C for not less than 5 hours. Thereafter, the dried pellets were injection-molded at a resin temperature of 270°C and a mold temperature of 80°C to prepare a test specimen having a diameter of 50 mmØ and a thickness of 3 mm.

The thus-prepared test specimens were exposed to 25 kGy of a cobalt-60 gamma radiation and then they were measured the change in yellowness index (Δ YI) thereof. The measurements of the yellowness index of the test specimens were carried out according to JIS K7103 by using a color difference meter (SM-3-CH, manufactured by SUGA Test Instruments Co., Ltd). The results are also shown in Table 5.

Table 5

40	
45	

35

Everyl, M	Tame 3		
Example No.	Aromatic compound containing oxy group or carbonyl group/compound containing sulfonate group	Blending ratio	Δ
Example 20	dibenzyl ketone/phenyl toluene-sulfonate	(part by weight)	
Example 21	dibenzyl ether/methyl toluene-sulfonate	0.5/0.5	9.
Example 22	dibenzyl ether/2-methoxyethyl toluene-sulfonate	0.25/0.25	8.
Example 23	benzyl benzoato/methyl toluene-sulfonate	0.5/0.5	3.
Example 24	benzyl benzoate/methyl toluene-sultonate	0.5/1.0	9.
Example 25	1,2-dibenzyloxy ethane/2-methoxyethyl toluene-sulfonate	0.5/0.5	4.3
Example 26	dicyclohexyl phthalate/phenyl toluene-sulfonate	0.5/0.75	8.8
Comparative Example 8	dibenzoyl methane/1,2-bis-tosyloxy ethane	0.5/0.5	9.3
	-/methyl toluene-sulfonate	-/0.5	14.

50

Examples 27 to 33 and Comparative Example 9:

An aromatic compound containing oxy group or carbonyl group and an aromatic hydrocarbon-aldehyde resin both shown in Table 6 were blended, at blending ratios also shown in Table 6, with 100 parts by weight of a polycarbonate resin in a tumbler. The mixture was fed into a vented single-screw extruder having a screw diameter of 40 mmØ and

Specifically, the raw materials used in this example were as follows.

(1) Polycarbonate resin:

lupiron S-2000 having a viscosity-average molecular weight of 25,000, produced by Mitsubishi Engineering Plastics Co., Ltd. (2) Aromatic hydrocarbon-aldehyde resin:

NIKANOL DS produced by Mitsubishi Gas Chemical Co., Ltd., from which no acetal group was detected; 5 NIKANOL L produced by Mitsubishi Gas Chemical Co., Ltd. and having an oxygen content of 10 % by weight; or NIKANOL Y-50 produced by Mitsubishi Gas Chemical Co., Ltd. and having an oxygen content of 18 % by weight.

The thus-prepared pellets were dried in a hot-air drier at a temperature of 120°C for not less than 5 hours. Thereafter, the dried pellets were injection-molded at a resin temperature of 270°C and a mold temperature of 80°C to prepare a test specimen having a diameter of 50 mmØ and a thickness of 3 mm.

The thus-prepared test specimens were exposed to 25 kGy of a cobalt-60 gamma radiation and then they were measured the change in yellowness index (Δ YI) thereof. The measurements of the yellowness index of the test specimens were carried out according to JIS K7103 by using a color difference meter (SM-3-CH, manufactured by SUGA Test Instruments Co., Ltd). The results are also shown in Table 6.

Example No.	Table 6		
	Aromatic compound containing oxy group or carbonyl group/aromatic bydross t	Plant	_
Example 27	carbonyl group/aromatic hydrocarbon-aldehyde resin dibenzyl ether/NIKANOL-Y-50	Blending ratio (part by weight)	1
Example 28	dibenzyl ketone/NIKANOL-Y-50	0.5/0.5	+
Example 29	1,4-bis-methowmeath	0.5/0.5	-
Example 30	1,4-bis-methoxymethyl benzene/NIKANOL-Y-50	0.5/0.5	-
Example 31	1,2-dibenzyloxy ethane/NIKANOL-Y-50	0.5/0.5	8
Example 32	hydroquinone-dibenzyl ether/NIKANOL-L benzyl benzoate/NIKANOL-L	0.5/0.75	
Example 33	dibenzoyl methane/NIKANOL-DS	0.25/0.5	.8 9.
Comparative Example 9	-/NIKANOL-DS	0.5/0.25	9.
		-/1.0	10.

35

50

30

20

25

Examples 34 to 39 and Comparative Example 10:

An aromatic compound containing oxy group or carbonyl group, a compound containing cyclic acetal group and a polyalkylene glycol derivative shown in Table 7 were blended, at blending ratios also shown in Table 7, with 100 parts by weight of a polycarbonate resin in a tumbler. The mixture was fed into a vented single-screw extruder having a screw diameter of 40 mmØ and extruded into pellets at a barrel temperature of 270°C. Specifically, the raw materials used in this example were as follows.

45 (1) Polycarbonate resin:

lupiron S-2000 having a viscosity-average molecular weight of 25,000, produced by Mitsubishi Engineering Plas-(2) Polyalkylene glycol derivative:

PEG1000: polyethylene glycol having an average molecular weight of 1,000.

PPG2000: polypropylene glycol having an average molecular weight of 2,000, or

PPGST30: polypropylene glycol-distearate having an average molecular weight of 3,000.

The pellets were then dried in a hot-air drier at a temperature of 120°C for not less than 5 hours. Thereafter, the dried pellets were injection-molded at a resin temperature of 270°C and a mold temperature of 80°C to prepare a test

The thus-prepared test specimens were exposed to 25 kGy of a cobalt-60 gamma radiation and then they were measured the change in yellowness index (Δ YI) thereof. The measurements of the yellowness index of the test specimens were carried out according to JIS K7103 by using a color difference meter (SM-3-CH, manufactured by SUGA

Furthermore, the dried pellets prepared above were injection-molded at a resin temperature of 270°C and a mold temperature of 80°C under an internal mold pressure of 550 Kg/cm² to form a cup-like molded product. The molded product was removed from the mold to evaluate mold-release properties thereof and measure a mold release resistance (kg) thereof. The results are also shown in Table 7.

10

. 15

20

25

30

35

Table 7

Example No.	Aramatia samuel	T	,	•	
	Aromatic compound containing oxy group or carbonyl group/compound containing cyclic acetal group/polyalkylene glycol derivative	Blending ratio (part by weight)	ΔΥΙ	Mold-release properties	Mold release resista nce (Ko
Example 34	benzyl benzoate/2-ben- zyl-1,3-diox- olane/PEG1000	0.25/0.25/0.25	9.4	good	492
Example 35	dibenzyl ether/2-benzyl- 1,3-dioxolane/PPG2000	0.2/0.2/0.2	8.3	good ·	503
Example 36	benzoin/2,2'-trimethylene bis-1,3-diox- olane/PPGST30	0.25/0.5/0.5	7.8	good	435
Example 37	dibenzyl ether/2-methoxy- 1,3-dioxolane/PPG2000	0.5/0.5/0.25	8.7	good	490
Example 38	1,2-dibenzyloxy ethane/2- benzyl-4-methyl-1,3-diox- olane/PPGST30	0.5/0.25/0.25	8.2	good	488
Example 39	dicyclohexyl phthalate/4- phenyl-1,3-diox- ane/PEG1000	0.2/0.2/0.5	9.2	good	439
Comparative Example 10	Not added	-	18.1	bad	810

Claims

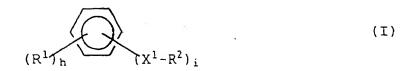
1. A polycarbonate resin composition comprising:

100 parts by weight of a polycarbonate resin;

0.01 to 5 parts by weight of an aromatic compound containing oxy group or carbonyl group; and

0.01 to 5 parts by weight of at least one compound selected from the group consisting of:

- (A) a compound containing cyclic acetal group;
- (B) a sulfone compound;
- (C) a sulfoxide compound;
- (D) polyalkylene glycol, an ether of polyalkylene glycol or an ester of polyalkylene glycol;
- (E) a compound containing sulfonate group; and
- (F) an aromatic hydrocarbon-aldehyde resin.
- 2. The polycarbonate resin composition according to claim 1, wherein said aromatic compound containing oxy group or carbonyl group is represented by the following general formula (I) or (II):



20

25

30

35

50

55

$$(R^3)_{j} \qquad (II)$$

where R¹, R³ and R⁵ are independently a (C₁-C₁₀)alkyl group, a (C₁-C₁₀)alkoxy group, a (C₁-C₁₀)alkoxy(C₁- C_{10})alkyl group, a (C_6-C_{16}) aryl (C_1-C_{10}) alkoxy (C_1-C_{10}) alkyl group, -CH₂OH or a halogen atom; R⁴ is a (C_1-C_1) C_{15})alkylene group, a (C_2 - C_{15})alkenylene group, a (C_6 - C_{30})arylene group, a (C_1 - C_{15})alkylene-dioxy group, or a (C₆-C₃₀)arylene-di(C₁-C₁₅)alkylene-oxy group, in which the said arylene group or the said arylene-dialkylene-oxy group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C1-C₁₀)alkyl group or a halogen atom; X¹, X² and X³ are independently -O-, -CHR⁶-O-, -CO-, -CHR⁷-CO-, -CO-COor -CO-C(OR8)2-; R2 R6, R7 and R8 are independently a hydrogen atom, a (C1-C30)alkyl group, a (C3-C₃₀)cycloalkyl group, a (C₂-C₃₀)alkenyl group, a (C₆-C₃₀)aryl group, a (C₆-C₃₀)aryl (C₁-C₃₀)alkyl group, a (C₆-C₃₀)aryl group, C_{30})aryl(C_2 - C_{30})alkenyl group, a hydroxy group, a (C_1 - C_{30})alkoxy group, a (C_6 - C_{30})aryl(C_1 - C_{30})alkoxy group, a (C_6-C_{30}) aryl (C_1-C_{30}) alkyl (C_1-C_{30}) alkoxy group, a (C_1-C_{30}) acyl group or a (C_6-C_{30}) aryl (C_1-C_{30}) acyl group in which the said aryl group, the said arylalkyl group, the said arylalkenyl group, the said arylalkoxy group, the said arylalkylalkoxy group or the said arylacyl group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C1-C10)alkyl group or a halogen atom; and h, j and k are independently an integer of 0 to 5 and i is an integer of 1 to 6 with provided that a sum of h and i is in the range of 1 to 6, and if i is an integer not less than 2, the R2's contained in the substituent groups bonded to the same aromatic ring may be covalently bonded with each other.

The polycarbonate resin composition according to claim 1, wherein said compound containing cyclic acetal group is represented by the following formula (III) or (IV):

$$(R^9R^{10}C)_p$$
 HC $(X^4)_1$ $R^{11}(X^5)_m$ CH $(CR^{12}R^{13})_q$ (III)

$$(R^{14}R^{15}C)_x HC - (X^6)_n R^{16}$$
 (IV)

where R^9 , R^{10} , R^{12} , R^{13} , R^{14} and R^{15} are independently a hydrogen atom, a (C_1-C_{10}) alkyl group, $-CH_2OH$, $-COOCH_3$, a phenyl group or a halogen atom; R^{11} is a (C_1-C_{15}) alkylene group, a (C_2-C_{15}) alkenylene group or a (C_6-C_{30}) arylene group, in which the said arylene group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C_1-C_{10}) alkyl group or a halogen atom; R^{16} is a hydrogen atom, a (C_1-C_{30}) alkyl group, a (C_3-C_{30}) cycloalkyl group, a (C_2-C_{30}) alkenyl group, a (C_6-C_{30}) aryl (C_1-C_{30}) alkenyl group, a (C_6-C_{30}) aryl (C_1-C_{30}) alkyl group, a (C_6-C_{30}) aryl (C_1-C_{30}) alkyl group, a halogen atom, a halogenated (C_1-C_{30}) alkyl group, an amino group or an amino

$$R^{27} S - R^{28}$$

$$\parallel$$

$$O$$
(VIII)

$$R^{29} = S - (R^{30} - S) - R^{31}$$
O
O
(IX)

15 .

35

where R^{27} , R^{28} , R^{29} , R^{31} , R^{32} and R^{35} are independently a (C_1 - C_{30})alkyl group, a (C_3 - C_{30})cycloalkyl group, a (C_2 - C_{30}) C_{30})alkenyl group, a (C_6 - C_{30})aryl group, a (C_6 - C_{30})aryl(C_1 - C_{30})alkyl group, a (C_6 - C_{30})aryl(C_2 - C_{30})alkenyl group, a (C_1-C_{30}) acyl (C_1-C_{30}) alkyl group, a (C_1-C_{30}) alkoxy (C_1-C_{30}) alkyl group, a (C_6-C_{30}) aryl (C_1-C_{30}) alkoxy (C_1-C_{30}) alkyl group, a (C_6-C_{30}) aryl (C_1-C_{30}) alkyl group, a (C_6-C_{30}) alkyl group, a $(C_6-C_$ group, a (C_6-C_{30}) aryl (C_1-C_{30}) alkoxy (C_2-C_{30}) alkenyl group, a (C_1-C_{30}) alkoxy group, a (C_1-C_{30}) alk group, a polyethylene-glycol-ether group or a pyridyl group, in which the said aryl group, the said arylalkyl group, the said arylalkenyl group, the said arylalkoxyalkenyl group and the said arylalkoxyalkyl group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C₁-C₄)alkyl group, a halogen atom, -NO₂, -N(R³⁶)₂, -COOH, -COOCH₃, -OH, -CH₂OR³⁷ or -OCH₃, and in which the said acylalkyl group, the said alkoxyalkyl group, the said arylalkoxyalkenyl group and the said arylalkoxyalkyl group may have substituent group(s) bonded to the alkyl or alkenyl chain thereof, the substituent group(s) being a (C1-C30)acyl group, a (C1- C_{30})alkoxy group or a (C_6 - C_{30})aryl(C_1 - C_{30})alkoxy group; R^{27} and R^{28} , R^{29} and R^{31} or R^{32} and R^{35} may be covalently bonded with each other; R^{30} , R^{33} and R^{34} are independently a (C_1-C_{15}) alkylene group, a (C_2-C_{15}) alkenylene group, a (C₆-C₃₀)arylene group, a (C₆-C₃₀)arylenedi(C₁-C₁₅)alkylene group, or -O(CH₂CHR³⁸O)₁-, in which the said arylene group and the said arylenedialkylene group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C_1-C_4) alkyl group, a halogen atom, -NO₂, -N(R³⁶)₂, -COOH, -COOCH₃, -OH, -CH₂OR³⁷ or -OCH₃, and R³⁸ are different from each other; R³⁶, R³⁷ and R³⁸ are independently a hydrogen atom, a (C_1-C_{10}) alkyl group, a (C_6-C_{15}) aryl group or a (C_6-C_{15}) aryl (C_1-C_{10}) alkyl group; and b, c and t are independently an integer of 1 to 100.

6. The polycarbonate resin composition according to claim 1, wherein said polyalkylene glycol, said ether of polyalkylene glycol or said ester of polyalkylene glycol is represented by the general formula (XI) or (XII):

$$R^{39}O - \{ (CH_2CH) - O\} - R^{41}$$
 (XI)

$$R^{42}COO - \{ (CH_2CH) - O \}_g - COR^{44}$$
 (XII)

where R^{39} , R^{41} , R^{42} and R^{44} are independently a hydrogen atom, a $(C_1\text{-}C_{30})$ alkyl group, a $(C_3\text{-}C_{30})$ cycloalkyl group, a $(C_2\text{-}C_{30})$ alkenyl group, a $(C_6\text{-}C_{30})$ aryl group, a $(C_6\text{-}C_{30})$ alkyl group or a $(C_6\text{-}C_{30})$ aryl $(C_2\text{-}C_{30})$ alkenyl group, in which the said aryl group, the said arylalkyl group and the said arylalkenyl group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a $(C_1\text{-}C_{10})$ alkyl group or a halogen atom; R^{40} and R^{43} are independently a hydrogen atom or a $(C_1\text{-}C_4)$ alkyl group; e and g are independently an integer of not less than 1, preferably 1 to 1000, and d and f are independently an integer of 1 to 10.

nated (C_1 - C_{30})alkyl group, in which the said aryl group, the said arylalkyl group, the said arylalkyl group or the said arylalkyl group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a (C_1 - C_{10})alkyl group or a halogen atom; X^4 , X^5 and X^6 are independently -CHR¹⁷-, -O-, -NH- or -PH- where R¹⁷ is a hydrogen atom, a(C_1 - C_{15})alkyl group or a (C_6 - C_{30})aryl group; and I, m and n are independently an integer of 0 or 1; and p, q and r are independently an integer of 1 to 10, and if p, q or r is not less than 2, the two or more of each of R⁹, R¹⁰, R¹² and R¹³ in the general formula (III), and R¹⁴ and R¹⁵ in the general formula (IV) may be the same or different.

4. The polycarbonate resin composition according to claim 1, wherein said sulfone compound is represented by the following formula (V), (VI) or (VII):

$$\begin{array}{c}
O \\
\parallel \\
S - R^{19} \\
\parallel \\
O
\end{array} (V)$$

where R^{18} , R^{19} , R^{20} and R^{22} are independently a $(C_1 - C_{30})$ alkyl group, a $(C_3 - C_{30})$ cycloalkyl group, a $(C_2 - C_{30})$ alkenyl group, a $(C_6 - C_{30})$ aryl group, a $(C_6 - C_{30})$ aryl group, a $(C_6 - C_{30})$ aryl group, a $(C_1 - C_{30})$ alkyl group, a $(C_6 - C_{30})$ aryl $(C_1 - C_{30})$ alkoxy $(C_1 - C_{30})$ alkoxy $(C_1 - C_{30})$ alkoxy $(C_1 - C_{30})$ alkoxy $(C_1 - C_{30})$ alkyl group, or a $(C_6 - C_{30})$ aryl $(C_1 - C_{30})$ alkoxy $(C_2 - C_{30})$ alkenyl group, in which the said aryl group, the said arylalkoxyalkyl group and the said arylalkoxyalkenyl group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a $(C_1 - C_4)$ alkyl group, a halogen atom, -NO₂. -N(R^{25})₂. -OH, -CH₂OR²⁶ or -OCH₃ where R^{25} and R^{26} are independently a hydrogen atom, a $(C_1 - C_{10})$ alkyl group, a $(C_6 - C_{30})$ aryl group or a $(C_6 - C_{30})$ aryl $(C_1 - C_{10})$ alkyl group, and in which the said acylalkyl group, the said alkoxyalkyl group, the said arylalkoxyalkyl group, and the said arylalkoxyalkenyl group may have substituent group(s) bonded to its alkyl or alkenyl chain and selected from a $(C_1 - C_{30})$ acyl group, a $(C_1 - C_{30})$ alkoxy group, and $(C_1 -$

5. The polycarbonate resin composition according to claim 1, wherein said sulfoxide compound is represented by the following formula (VIII), (IX) or (X):

50

5

25

30

EP 0 753 540 A2

0.01 to 5 parts by weight of a compound containing cyclic acetal group; and0.01 to 5 % by weight of polyalkylene glycol, an ether of polyalkylene glycol or an ester of polyalkylene glycol.

 The polycarbonate resin composition according to claim 1, wherein said compound containing sulfonate group is represented by the following formula (XIII), (XIV) or (XV):

$$(R^{45})_{s} \downarrow 0 \qquad (XIII)$$

$$(S - OR^{46})_{u}$$

$$O$$

$$(R_{47})_{v} \xrightarrow{0} R^{48} \xrightarrow{0}_{w} R^{49}$$

$$(XIV)$$

$$(R_{50})_{x} \xrightarrow{0} (R^{51} - 0) \xrightarrow{y} \xrightarrow{g} (XV)$$

where R^{45} , R^{47} , R^{50} and R^{52} are independently a $(C_1\text{-}C_{30})$ alkyl group, a $(C_6\text{-}C_{30})$ aryl group, a $(C_6\text{-}C_{30})$ arylsulfone group or a halogen atom; R^{46} and R^{49} are independently a hydrogen atom, a $(C_1\text{-}C_{30})$ alkyl group, a $(C_3\text{-}C_{30})$ cycloalkyl group, a $(C_2\text{-}C_{30})$ alkenyl group, a $(C_6\text{-}C_{30})$ aryl group, a $(C_6\text{-}C_{30})$ aryl $(C_1\text{-}C_{30})$ alkenyl group or a $(C_1\text{-}C_{30})$ acyl group, in which the said aryl group, the said arylalkyl group and the said arylalkenyl group may have substituent group(s) bonded to the aromatic ring thereof, the substituent group(s) being a $(C_1\text{-}C_4)$ alkyl group or a halogen atom; R^{48} and R^{51} are independently a $(C_1\text{-}C_{15})$ alkylene group, group(s) bonded to the aromatic ring thereof, the substituent group(s) being a $(C_1\text{-}C_4)$ alkyl group or a halogen atom; s, v, x and z are independently an integer of 0 to 5 and u is an integer of 1 to 6, preferably 1 to 3 with provided from 1 to 500.

- 40 8. The polycarbonate resin composition according to claim 1, wherein said aromatic hydrocarbon-aldehyde resin is prepared by reacting at least one aromatic hydrocarbon selected from the group consisting of benzene, toluene, naphthalene, methyl naphthalene, ethyl naphthalene, dimethyl benzene, tetramethyl benzene, pseudo-cumene, cumene, naphthalene, methyl naphthalene, ethyl naphthalene, dimethyl naphthalene, acenaphthene and anthracene with at hyde, iso-butylaldehyde, valeraldehyde, laurinaldehyde, stearinaldehyde, glyoxal, succinclialdehyde, butylaldetonaldehyde, propiolaldehyde, benzaldehyde, tolylaldehyde, salicylaldehyde, cinnamaldehyde, naphthaldehyde, tufurlural, methylal, dioxolane, trioxane, tetraoxane, paraformaldehyde, paraldehyde and metaldehyde, in the presence of an acid catalyst.
- The polycarbonate resin composition according to claim 1, wherein said aromatic hydrocarbon-aldehyde resin contains substantially no acetal group.
 - The polycarbonate resin composition according to claim 1, wherein said aromatic hydrocarbon-aldehyde resin has an oxygen content of at least 8 % by weight.
 - 11. A polycarbonate resin composition comprising:

5

10

15

20

25

30

35

55

100 parts by weight of a polycarbonate resin;

0.01 to 5 parts by weight of an aromatic compound containing oxy group or carbonyl group;



EUROPEAN SEARCH REPORT

Application Number

EP 96 11 1313

ategory	Citation of document with ind of relevant passa		Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.CI.6)
· ·	US 5 214 078 A (LUND 25 May 1993 * examples 4,5 *	Y CHARLES E ET AL)	1	C08L69/00 C08K5/00
X	EP 0 359 366 A (MITS 21 March 1990 * claims 1,5; exampl		1	
X	EP 0 518 149 A (BAYE * claim 3 *	R AG) 16 December 1992	1	
X	EP 0 525 338 A (MILE * claims 1,7 *	S INC) 3 February 1993	1.	
X	EP 0 535 464 A (BAYE * claim 1 *	ER AG) 7 April 1993	1.	
X	EP 0 561 238 A (BAYE 22 September 1993 * example 3 *	ER AG)	1	TECHNICAL FIELDS
X .	EP 0 637 609 A (TEIX 8 February 1995 * claim 1 *	JIN CHEMICALS LTD)	1	TECHNICAL FIELDS SEARCHED (Int.CI.6) CO8K CO8L
χ	EP 0 640 646 A (TEIO * table 1 *	JIN LTD) 1 March 1995	1	
X	PATENT ABSTRACTS OF vol. 016, no. 152 (6 & JP 04 007356 A (6 10 January 1992 * abstract *	JAPAN C-0929), 15 April 1992 MITSUBISHI KASEI CORP)	1	
 - 	,	-/		
·				
		hoop drown up for all plains		
	The present search report has I	Ogte of completion of the search	1	Exertiner
	THE HAGUE	29 June 1999	Sc	chueler, D
Y:p	CATEGORY OF CITED DOCUMENTS articularly relevant if taken alone articularly relevant if combined with anot accomment of the same category schnological background	T : theory or princ E : earlier patent of after the filling ther D : document cite L : document cited	ple underlying the locument, but putate d in the applicable	ne invention zilished on, or on



Europäisches Patentamt

European Patent Office

Office européen des brevets



(11) EP 0 753 540 A3

(12)

EUROPEAN PATENT APPLICATION

(88) Date of publication A3: 25.08.1999 Bulletin 1999/34

(51) Int. Cl.6: C08L 69/00, C08K 5/00

(43) Date of publication A2:15.01.1997 Bulletin 1997/03

(21) Application number: 96111313.1

(22) Date of filing: 12.07.1996

(84) Designated Contracting States: BE DE FR

(30) Priority: 12.07.1995 JP 17624895 26.07.1995 JP 19065795 20.09.1995 JP 24154695 28.11.1995 JP 30921195 28.11.1995 JP 30921295 23.02.1996 JP 3634496 19.03.1996 JP 6261596

(71) Applicant: MITSUBISHI ENGINEERING-PLASTICS CORPORATION Tokyo 104 (JP)

(72) Inventors:

Miya, Shinya,
 c/o Mitsubishi Eng.-Plastics Corp.
 Hiratsuka-shi, Kanagawa-ken (JP)

Kanayama, Satoshi,
 c/o Mitsubishi Eng.-Plast. Corp
 Hiratsuka-shi, Kanagawa-ken (JP)

Shimomai, Ken,
 c/o Mitsubishi Eng.-Plastics Corp.
 Hiratsuka-shi, Kanagawa-ken (JP)

(74) Representative:
Hansen, Bernd, Dr. Dipl.-Chem. et al
Hoffmann Eitle,
Patent- und Rechtsanwälte,
Arabellastrasse 4
81925 München (DE)

(54) Polycarbonate resin composition

(57) A polycarbonate resin composition according to the present invention is suited to be used for medical supplies or appliances and shows a very low yellow discoloration when exposed to an ionizing radiation for sterilization. The polycarbonate resin composition comprises 100 parts by weight of a polycarbonate resin; 0.01 to 5 parts by weight of an aromatic compound containing oxy group or carbonyl group; and 0.01 to 5 parts by weight of at least one compound selected from the group consisting of: (A) a compound containing cyclic acetal group; (B) a sulfone compound; (C) a sulfoxide compound; (D) polyalkylene glycol, an ether of polyalkylene glycol or an ester of polyalkylene glycol; (E) a compound containing sulfonate group; and (F) an aromatic hydrocarbon-aldehyde resin.



EUROPEAN SEARCH REPORT

Application Number

EP 96 11 1313

		IDERED TO BE RELEVANT	•	
Category	Citation of document wi of relevant p	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.CI.6)	
X	PATENT ABSTRACTS vol. 016, no. 164		1	AFFECATION (Int.Cl.6)
-	PATENT ABSTRACTS vol. 018, no. 615 24 November 1994 & JP 06 234910 A 23 August 1994 * abstract *	OF JAPAN (C-1277), (JAPAN ENERGY CORP),	1	
				
	•			
				TECHNICAL FIELDS SEARCHED (Int.CI.6)
				SEARCHED (Int.Cl.6)
				•
	•	•		·
		•		
		• •		
-	· , · ·			
				•
	The present search report has	been drawn up for all claims	1	
F	Place of search	- Date of completion of the search	- 	Examiner
Ţ	HE HAGUE	29 June 1999	Schue	eler, D
: particul : particul docum	EGORY OF CITED DOCUMENTS larly relevant if taken alone larly relevant if combined with ance ent of the same category logical background	E : earlier patent do	le underlying the inver- cument, but published	notion .

FORM 1503 03.82 /Po4CA

ANNEX TO THE EUROPEAN SEARCH REPORT ON EUROPEAN PATENT APPLICATION NO.

EP 96 11 1313

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

29-06-1999

Patent document cited in search repr		Publication date		Patent family member(s)	Publication date
US 5214078	A	25-05-1993	CA	2097374 A	04-12-19
			EP	0572889 A	08-12-19
			JP	6093192 A	05-04-19
			US	5382605 A	17-01-19
EP 0359366	Α	21-03-1990	JP	2071752 A	12-03-19
			JP	6002156 B	12-01-19
			JP	2129261 A	17-05-19
			JP	7068446 B	26-07-19
			CA	1338093 A	27-02-19
			DE	68913633 D	14-04-19
			DE	68913633 T	09-06-19
			KR	9701500 B	11-02-19
EP 0518149	Α	16-12-1992	DE	4119329 A	17-12-19
			JP	5179127 A	20-07-19
			US	5476893 A	19-12-19
EP 0525338	Α	03-02-1993	CA	2069595 A	21-12-19
			US	5280050 A	18-01-19
EP 0535464	Α	07-04-1993	DE	4132629 A	08-04-19
			DE	59205462 D	04-04-19
		•	JP	5209120 A	20-08-19
			US	5274009 A	28-12-19
EP 0561238	Α	22-09-1993	DE	4221581 A	23-09-19
			JP .	6041413 A	15-02-19
*********			US	5326800 A	05-07-19
EP 0637609	·A	08-02-1995	JP	7041653 A	10-02-19
		•	CA	2128823 A	28-01-19
•			DE	69415096 D	21-01-19
			US	5478874 A	26-12-19
EP 0640646	Α	01-03-1995	JP	7062077 A	07-03-19
			JР	7126374 A	16-05-19
			JР	7165905 A	27-06-19
			US	5486555 A	23-01-19
		•	US	5494952 A	27-02-19
		•	US	5668202 A	16-09-19

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82